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ABSTRACT OF RESEARCH PROGRESS

Substantial progress has been made in processing nano-heterogeneous, amorphous thin films with attractive magnetic properties. Work has been concentrated on two categories of sputtered thin films; (1) soft magnetic, CoB and CoNbZr alloy thin films, and (2) magneto-optical, compositionally modulated Gd/Co multilayer thin films. We are attempting to understand and control the nanoscale heterogeneous structures of these films for application to, respectively, thin film magnetic recording heads, and magneto-optical recording. Cobalt, Niobium, Zirconium, Boron, [illegible])

For the first category, the major results obtained thus far are: (1) the fabrication of soft magnetic thin films with state of the art figures of merit for recording head applications. CoB and CoFeB films with initial permeabilities of 5000-10,000 have been produced by annealing in rotating magnetic fields. The best films have a 3 dB rolloff frequency in excess of 100 MHz. The complex permeability spectra of soft amorphous thin films have been resolved for the first time in this work.

In the second category, sequentially sputtered, compositionally modulated films CMF with an average composition of $\langle Gd_{25}Co_{75} \rangle$ have been prepared with bilayer periods from 1-15 nm. We have studied the as-deposited, annealed, and ion-beam mixed CMF by AES, TEM/TED, X-ray, Lorentz microscopy, magnetometry, and measured their Kerr magneto-optic coefficients. The films have an interesting nanoscale morphology, but little vertical anisotropy, and small Kerr rotations. The research has produced considerable insight to the role of included oxygen on film chemical and structural stability. Investigations of GdFeCo, TbFe, and TbFeCo CMF films are in progress.

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MAGNETIC PROPERTIES OF NANO-HETEROGENEOUS AMORPHOUS THIN FILMS

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I. OBJECTIVES OF THE RESEARCH

The overall goal of this research was to study the fabrication and properties of nano-heterogeneous, compositionally modulated, amorphous thin films with attractive properties for magnetic device applications. Devices of interest included; magnetic and magneto-optic recording media, thin film recording heads, and magnetic sensors.

The first year of the program study was to be concentrated on the processing and characterization of sputtered films processed by a variety of techniques that produced controllable nanoheterogeneity. The primary tasks to be completed in the first year included:

1. Study the effect of sputter deposition parameters on; (a) film nanostructure as determined by TEM, and average composition, (b) temperature dependent magnetization and magnetic hysteresis, (c) temperature and frequency dependent permeabilities and permitivities, (d) film morphologies, and (e) film magnetization distributions.
2. Study of the effect of post deposition annealing to optimize the film magnetic permeability and resistivity, and to minimize their magnetic anisotropy.
3. Study the correlation of process variables and film nanostructure, and their effect on magnetic and electronic properties.

II. STATUS OF THE RESEARCH EFFORT

In this first year of the program we have made substantial progress toward the goal of processing nano-heterogeneous, compositionally modulated, amorphous thin films with attractive magnetic properties. We have concentrated our work on two categories of sputtered thin films; (1) soft magnetic thin films of Co-based alloys, and (2) compositionally-modulated multi-layer films of rare earth, and transition metals. The overall goal was to study the possibility of enhancing the magnetic response of the films in each category by understanding, and controlling their nanoscale features and morphology. In the first category of soft films we stressed the possibility of maximizing the product of the RF initial permeability and the resistivity as required in applications, such as integrated thin film magnetic recording

heads, where a large high frequency response is required. In the second category we were concerned with maximizing the magneto-optical response of amorphous thin films for use in applications such as magneto-optic recording. In the following we will separately summarize the progress made in each of these areas.

MAGNETICALLY SOFT, NANO-HETEROGENEOUS, AMORPHOUS THIN FILMS

We have fabricated nano-heterogeneous thin films by two sputtering techniques.

In the first, Co-base alloy films were sequentially RF-diode sputtered from simultaneously excited elemental targets. Thin films with Co-B and Co-Zr binary, and Co-Fe-B ternary, compositions were deposited. This work took advantage of a considerable amount of work in progress from a previous contract (F33657-84-C-2058), but was completed as part of the first year effort on this program. The large portion of this research was devoted to a systematic study of RF sequentially sputtered thin films. This work was completed with support from this program and is largely summarized in a recent Ph.D. dissertation by Dr. Dae Yong Kim of our laboratory [1].

The major results of this work are as follows:

1. TEM and related studies revealed that the major source of the anisotropy of the as-deposited soft magnetic thin films was due to growth anisotropies that resulted from self shadowing of obliquely incident atomic flux in the growing film. Our experiments conclusively show that this anisotropy can be virtually eliminated in films with a normally incident sputtered flux. The resultant films are still semi-hard (H is a few oersteds) and have unacceptably low initial permeabilities (less than 1000); likely due to the presence of a perpendicular component of the magnetization.

2. We produced very soft thin films of Co-B and Co-Fe-B with high initial permeabilities of 5,000-10,000 by annealing at elevated temperatures in rotating magnetic fields. On the other hand, simple thermal annealing was not particularly effective in reducing the anisotropy of the as-deposited films. The best binary Co-B films had permeabilities greater than 6000 with a 3 dB roll-off at 30 MHz, and the best ternary Co-Fe-B films had permeabilities of greater than 10,000 and a 3 dB roll-off at 100 MHz. The binary films were essentially isotropic but the ternary films had anisotropy of approximately 1.40 Oe. These values of high frequency permeabilities are, we believe, the

highest ever reported in the literature. The data was acquired with a new state-of-the-art thin film permeameter system developed in our lab and described in a recent journal article [2]. This system resolved the complete, swept, complex permeability spectrum of amorphous films for the first time, and the results have led to some unique observations to be discussed.

3. Using a combination of TEM and AES we have shown, for the first time, the relationship between film nanoscale features and morphologies produced by the important film deposition, and post-deposition, parameters on the in-plane anisotropies of magnetically soft films. These studies conclusively show that, in the as-deposited films, the major anisotropy is a dipolar anisotropy associated with the shape demagnetizing fields of the nanoscale features in the film morphology. At present we are utilizing image processing techniques to quantify these results in terms of the statistical characteristics of the nanoscale morphology. A paper has been recently published describing the results of these studies [4].

4. An important observation made in these studies was that of a greatly extended range of compositions in the Co-B binary system (relative to melt-quenched alloys) for which stable amorphous alloys could be retained in thin film form. Stable amorphous thin films with up to 40% boron could be sputtered, compared to a maximum of about 25% boron for melt-quenched ribbons. TEM revealed that sequentially sputtered films with an average composition of $<Co_{61}B_{39}>$ were near the percolation limit and consisted of two, co-existing amorphous phases with morphological features of 1-10 nm. Surprisingly these films responded well to rotating field annealing and had attractive magnetic properties. The details of these studies of anomalous films has been reported in a recent journal article [5].

5. We have also conducted the first extensive, systematic studies of the high frequency bias-susceptibilities of soft, nano-heterogeneous, amorphous thin films. Our results were recently reported in the J. of Applied Physics [6]. In these studies we made the first observation of completely reversible transverse susceptibilities produced in a $<Co_{61}B_{39}>$ binary alloy thin film by rotating field annealing. This data indicates that the internal field due to the magnetization ripple was larger than the anisotropy field opposing the rotation of the mean magnetization. We believe that this is the first observation of a ripple dominated response in any magnetic material.

6. In related studies we have been studying the origin of the dynamical response of the magnetization of these soft amorphous thin films. Our work shows, for the first time, that the complex initial susceptibility of these films exhibits a classical relaxation with no detectable resonant character. Computer simulations show that the response is that of a highly damped rotation. The origin of the damping is not understood at present, but appears to be associated with the magnetization ripple. This work has also been reported in a recent publication [6].

7. We have studied the magnetization distribution in these soft amorphous films with interesting properties by a combination of Bitter patterns, Kerr effect microscopy, and Lorentz microscopy. These works revealed that the films were nearly single domain. LEM of thinner samples revealed a significant magnetization ripple which was quantitatively analyzed, for the first time, by two dimensional, digital image processing. This work was presented at the European Magnetics Materials and Applications Conference (EMMA) in Salford, UK in Sept. 1987, where it received the Best Poster Award. This paper has recently been published in the IEEE Transactions on Magnetics [7].

The second technique to be used in this program for producing nanoheterogeneous magnetic thin films is magnetron sputtering. We will either reactively sputter alloy films, or utilize chip sputtering in the sputter-up mode of deposition, to deposit alloy films. We have recently installed and are operating a new multi-target magnetron sputtering system for this work. The new system has a vacuum load lock that will permit more stringent control over the impurities present during deposition. In addition to the unique sputter-up mode of deposition, it is equipped with a two gas mixing valve for simultaneously admitting the inert sputtering gas (Ar) and a reactive gas (O or N) to the deposition chamber. We have ordered and will install a residual gas analyzer and partial pressure feedback control system. The only work conducted in this system thus far was concerned with calibrating sputtering rates and analyzing film compositions. The system is equipped with a pure Co target (for use with boron oxide and nitride chips), a ternary Co-Nb-Zr target, and a quaternary Co-Fe-Si-B target. Early indications are that films with excellent soft magnetic properties will be obtained.

NANO-HETEROGENEOUS, COMPOSITIONALLY MODULATED (CMF), MAGNETO-OPTICAL THIN FILMS

There are considerable theoretical and experimental indications that the vertical anisotropy in amorphous thin films of rare earth-transition metal alloys is due to their nano-heterogeneity. The amount, and spatial distribution, of included oxygen and argon is believed to be particularly important in determining their magnetization distributions and anisotropies. Initially we are experimentally investigating these issues to systematically determine the influence of deposition and post deposition parameters on the vertical anisotropy in compositionally modulated Gd-Co thin films. This system was selected for initial study because the sputtering targets were on hand, and because of the wide amount of information available in the literature on these binary alloy films.

In the past eight months we conducted an extensive study of compositionally modulated Gd-Co thin films made by sequentially sputtering from the elemental targets. The average composition was fixed near $\langle Gd_{25}Co_{75} \rangle$ and the bilayer period was varied from 1-15 nanometers. These films were subsequently ion beam mixed by high dose implants of Ar ions to investigate the possibility of producing amorphous films with a greater vertical anisotropy. This method, rather than alloy sputtering, was chosen to obtain the amorphous films because of the possibility of realizing unique nano-heterogeneous films with enhanced Kerr magneto-optic coefficients. This possibility is thought to be associated with the coexistence of nanoscale optical birefringence and magneto-optic activity.

We have studied the as-deposited, annealed, and ion-beam mixed CMF by a combination of AES, TEM/TED, cross-section TEM, X-ray, Lorentz microscopy, Bitter patterns, magnetometry, and have measured their Kerr magneto-optic coefficients. A considerable volume of data has been obtained, but has been only partially analyzed as yet. The principal results obtained thus far are summarized below:

1. TEM revealed that all of the as-deposited CMF were polycrystalline, and that only the ion beam mixed CMF with the smallest bilayer periods (1 nm) were amorphous. The TEM nanostructure of the latter consisted of a porous network. Only the annealed films with the 1 nm period exhibited a vertical anisotropy as indicated by the development of domain wall coupling in Bitter patterns. On the other hand, annealing at 250 °C significantly increased the Kerr magneto-

optic rotation of films with bilayer periods as large as 10 nm. Although these results are not particularly surprising, the Kerr rotations obtained were quite small. In the next period we plan to repeat many of these experiments with CoFeTb alloy thin films which are known to have significantly larger vertical anisotropy and Kerr rotation.

2. The research conducted on the Gd-Co binary alloy films has produced considerable insight into the role of oxygen on their chemical and structural stability, and hence on their vertical anisotropy. In the experiments, oxygen incorporated into the films was systematically controlled by the application of an RF substrate bias during deposition and by post deposition annealed, and was monitored by AES. The results of this work have recently been published [7].

III. CUMULATIVE CHRONOLOGICAL LIST OF PUBLICATIONS

(1) D. Y. Kim, A Study of the Effect of Process-Parameters on the Magnetic Anisotropy of Cobalt-Based Soft Amorphous Thin Films, Ph.D. Dissertation, University of Texas at Austin, Spring 1988.

(2) C. Grimes, P.L. Trouilloud, and R. M. Walser, "A New Swept Frequency Thin Film Permeameter," IEEE Trans. Magn. MAG-24, 603 (1988).

(3) I. S. Jeong and R. M. Walser, "Quantitative Study of Magnetization Ripple in $Co_{61}B_{39}$ Thin Films," IEEE Trans. Magn. MAG-24, 1725 (1988).

(4) A. P. Valanju, I. S. Jeong, D. Y. Kim, and R. M. Walser, "Correlation of Process Parameters with Morphology and Magnetic Properties of Co-Based Amorphous, Soft, Thin Films," J. Appl. Physics 64, 5443 (1988).

(5) D. Y. Kim, I. S. Jeong, and R. M. Walser, "Anomalously Soft Magnetic Properties of Boron-Rich CoB Thin Films," J. Appl. Phys. 64, 5676 (1988).

(6) I. S. Jeong, A. P.Valanju, and R. M. Walser, "High Frequency Bias-Susceptibilities of Ultra-Soft CoB Amorphous Thin Films," J. Appl. Phys. 64, 5679 (1988).

(7) Geon Choe, M. F. Becker, and R. M. Walser, "Effect of Oxygen on Compositionally Modulated Gd-Co Thin Films," J. Appl. Phys. 64, 6104 (1988).

(8) D. Y. Kim and R. M. Walser, "Magnetization, Composition, and Magnetic Properties in Sequential RF Co-Sputtered $Co_{100-x}B_x$ Thin Films," paper to be prepared in Fall 1988.

(9) D. Y. Kim and R. M. Walser, "Magnetic and Structural Properties of Sputtered Amorphous $Co_{100-x}B_x$ (15 at% < x < 39 at%)," paper to be prepared in Fall 1988.

(10) I. S. Jeong, A Study of High Frequency Bias-Susceptibilities in Cobalt-Based Amorphous Magnetic Thin Films," Ph.D dissertation in preparation.

IV. PROFESSIONAL PERSONNEL ASSOCATIATED WITH THE RESEARCH

1. Professor Rodger M. Walser - Principal Investigator
2. Ms. Alaka P. Valanju - Programmer/Analyst
3. Dr. Dae Yon Kim - Research Associate (Ph.D Degree awarded-May 1988).
4. Mr. In-Seop Jeong - Graduate Research Associate - Ph.D. Student in Materials Science and Engineering.
5. Mr. Craig Grimes - Graduate Research Associate - Ph.D. Student in Electrical and Computer Engineering.
6. Mr. Geon Choe - Graduate Research Associate - Ph.D. Student in Materials Science and Engineering.
7. Mr. Ernest Louis - Graduate Research Associate - Ph.D. Student in Materials Science and Engineering.
8. Ms. Elizabeth Savage - Technical Secretary.

Advanced Degree Awarded:

1. Dr. Dae Yong Kim, Ph.D., May 1988, Thesis: A Study of the Effect of Process-Parameters on the Magnetic Anisotropy of Cobalt-Based Soft Amorphous Thin Films, Ph.D. Dissertation, University of Texas at Austin, Spring 1988.

V. PAPERS PRESENTED AT CONFERENCES, MEETINGS, ETC.

1. I. S. Jeong and R. M. Walser, "Quantitative Study of Magnetization Ripple in $Co_{61}B_{39}$ Thin Films," European Magnetic Materials and Applications Conference, Salford, U.K., Sept. 1987.
2. A. P. Valanju, I. S. Jeong, D. Y. Kim, and R. M. Walser, "Correlation of Process Parameters with Morphology and Magnetic Properties of Co-Based Amorphous, Soft, Thin Films," 4th Joint MMM-Intermag Conference, Vancouver, Canada, 1988.
3. D. Y. Kim, I. S. Jeong, and R. M. Walser, "Anomalously Soft Magnetic Properties of Boron-Rich CoB Thin Films," 4th Joint MMM-Intermag Conference, Vancouver, Canada, 1988.

4. I. S. Jeong, A. P.Valanju, and R. M. Walser, "High Frequency Bias-Susceptibilities of Ultra-Soft CoB Amorphous Thin Films," 4th Joint MMM-Intermag Conference, Vancouver, Canada, 1988.

5. Geon Choe, M. F. Becker, and R. M. Walser, "Effect of Oxygen on Compositionally Modulated Gd-Co Thin Films," 4th Joint MMM-Intermag Conference, Vancouver, Canada, 1988.